Attorney Docket No. P66718US0

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: ANTON et al.

Application No.: 09/857,181 Group Art Unit: 1772

Filed: June 19, 2001 Examiner: Patricia L. Nordmeyer

For: MICROPOROUS HEAT INSULATION BODY

SECOND DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

The undersigned Octavian ANTON does hereby declare and state that:

1. He is a named inventor of the subject application.

2. He attended University of Bucharest, Romania, Faculty of Geology and Mineralogy, first graduating therefrom in 1962, receiving therefrom the degree Ph.D. in Mineralogy in 1970, and at which he did post-doctorate graduate work in physico-chemical analytical techniques in 1971.

3. He worked from 1962 to 1964 with a geological prospection enterprise, from 1964 to 1972 as a researcher at the Geological Institute of the Romanian Academy of Science, and from 1972 to 1975 as a researcher at the Geological Institute of Romania involving clay mineralogy; in 1975 he became an Assistant at the Catholic University Louvain-la-Neuve, Belgium; and from 1976 to the present he has worked at Etex Group (formerly S.A. Eternit N.V.) – initially working in the field of mineral synthesis for applications in the building industry at the research center of S.A. Eternit N.V., in 1990 becoming Research and Development (R&D) Deputy Director, and currently holding the position Director, Promat

International Research and Technology Centre, Promat International NV (an Etex Group company), responsible for executing, supervising, and coordinating R&D activities in Belgium, England, and Austria for new products, and a new generation of products, in the fields of fire protection and high temperature insulation.

4. He is founder and president of the Romanian Clay Minerals Group and a member of the i.e., International Association for Clay Studies.

5. He is named inventor of a number of patents in the fields of fire protection, high synthetic mineral structures for application in chemical and bio-chemical catalysis, friction materials, and plastics.

6. In connection with the subject patent application, he had personally supervised research with the objective of overcoming weaknesses in commercially available super-insulators based on pyrogenic silica

7. He is familiar with the Office Action mailed December 2, 2002, including the rejection of claims under 35 USC 103(a), and the subsequent Advisory Action mailed September 3, 2003, maintaining the rejection under 35 USC 103(a), in the subject application and, in order to overcome the rejection, reports the following tests conducted and test results obtained under his direction and control:

The tested compositions:

- all percentages are by weight (w/w)

Invention Example 1 (dry mixture)

The dry mixture consists of 10% dry xonotlite (Promaxon®), 25% rutile (used as opacifier) - 3% silica-fibres- 62% pyrogenic silica.

The powders are mixed during 3 minutes mixed in a Kenwood mixer and then pressed at a density of 0.3g/cm3. The pressure applied was 1MPa.

Comparative Example 1a (wet mixture)

For the preparation of the wet mixture, the pyrogenic silica, silica-fibres and rutile are mixed as described above for the dry mixture. This powder mixture is then added to a slurry of xonotlite in water. The xonotlite-slurry used was the slurry normally produced for Promat Ca-silicate-based fire protection boards, slurry is similar to known xonotlite slurries used by industry to produce fire protection or high temperature insulation materials.

This wet mixture, which contains 50% dry phase and 50% water, is homogenized for 3 minutes and then pressed on a filter press applying a pressure of 1MPa.

Invention Example 2 (dry mixture)

5% Xonotlite powder, 3% silica-fibers, 25% ZrSiO4, and 67% pyrogenic silica are homogenously mixed by a planetary mixer (type Hobart). The resulting dry mixture is then put into a mold, and pressed to form a monolithic sample. The formed sample is easily demolded. It has a good surface aspect, no surface cracks, dimensional stability, and no drying shrinkage afterwards. Dry density of the sample is 350 g/cm3, and it has superior insulating properties. The sample material can be used as a product.

Comparative Example 2a (wet mixture)

As in Invention Example 1, 5% xonotlite powder, 3% silica-fibers, 25% ZrSiO4, and 67% pyrogenic silica are homogeneously mixed by a planetary mixer (type Hobart), but the xonolite is in an aqueous (xonolite) slurry (water content=89%). The amount of the dry mixture is kept the same as in Invention Example 1. The resulting wet mixture is then put into a mold and pressed to form a wet sample. After being dried at 60-105 oC, the sample shows slight drying shinkage, with visible micro cracks on its surface. Dry density of the sample is 400 g/cm3. The sample material cannot be used, directly, as a product.

Invention Example 3 (dry mixture)

40% Xonotlite powder, 3% silica-fibers, 25% ZrSiO4, and 67% pyrogenic silica are homogeneously mixed by a planetary mixer (type Hobart). The resulting dry mixture is then put into a mold, and pressed to form a monolithic sample. The formed sample is easily demolded. It has a good surface aspect, no surface cracks, dimensional stability, and no drying shrinkage afterwards. Dry density of the sample is 350 g/cm3, and it has superior insulating properties. The sample material can be used as a product (Appendix, Fig. 4 and Fig. 5).

Comparative Example 3a (wet mixture)

As in Invention Example 2, 40% xonotlite powder, 3% silica-fibers, 25% ZrSiO4, and 67% pyrogenic silica are homogeneously mixed by a planetary mixer (type Hobart), but the xonolite is in a (xonolite) slurry (water content=89%). The amount of the dry mixture is kept the same as in Invention Example 2. The resulting wet mixture is then put into a mold and pressed to form a wet sample. After being dried at 60-105 oC, the sample much drying shinkage and it has an inferior surface aspect; no dimension tolerance can be controlled (Appendix Fig. 4 and Fig. 5). Dry density of the sample material is 500 g/cm3; accordingly, it has a high thermal conductivity (=bad insulating properties). In addition, the material cannot be used, directly, as a product, and it needs to be sanded or cut to form a regular shape.)

The parameters examined

Integrity: the presence of cracks were checked by visual observation with a microscope and corresponding photographs taken, which are reproduced in the attached Fig. 1 and Fig. 2. The magnifications of the photographs in the attached Fig 1 and Fig. 2 are indicated by scale bars below the photographs.

Thermal insulation: Lambda measurements were taken according to the hot-wire-method with the KEM Model TC-51 High Temperature thermal conductivity meter (4 page meter brochure attached hereto). The measurements taken are recorded on the attached Fig. 3.

8. The tests and test results reported, herein, show that only the use of dry mixtures can lead to the production of a super-insulator. The wet method leads always to a standard type of insulator. The higher lambda values of the wet-processed system (Δ) relative to the dry system values (◊) recorded in Fig. 3 are due to destruction of the nanoporous silica structure by capillary forces on drying.

9. The destruction of nanoporous structure following wet processing of pyrogenic silica is a known phenomenon and, for this reason, the art taught using sub-critical drying in conjunction with wet processing for such kinds of silica structures (for example, xerogels).

10. The use of xonotlite as a powder in a microporous heat insulation body as described and claimed in the subject application is the result of detailed research into insulation characteristics, targeting:

    (i) Improvement of de-airing during pressing a dry, micro-nanoporous mixture for manufacturing a super-insulation body: It is known that such mixtures shaped by pressing develop a spring-back phenomenon during de-molding, which destabilizes the shaped product's matrix. Addition of the xonotlite (Promaxon®) in the invention example accordance with the teachings of the subject invention application , as demonstrated by the test reported herein, and as described in the subject patent

application provides control of this phenomenon. Previously the skilled person in the art solved this problem by sintering the product or by encapsulating the product in protective materials.

(ii) Improvement of bending strength: By eliminating the spring-back phenomenon, and due to the texture of xonotlite particles, bending strength is, also, improved for products such as super-insulators, which are known to be weak, difficult to handle, and to work with.

11. To achieve the aforesaid targeted insulation characteristics – i.e., those of a super-insulator – pyrogenic silica was found to be the best raw material. Pyrogenic silica is composed of nanoparticle aggregates having a nanoporous structure. The nanoporous structure is a key-parameter in the control of heat transfer through the finished super-insulator body.

12. When pressing a mixture containing pyrogenic silica, no contact with water is allowable, because the nanoporous structure is extremely sensitive and will be destroyed.

13. A salient feature of the xonotlite powder, in the invention described and claimed in the subject application, is the control of de-airing a pyrogenic-silica-based matrix during pressing in the manufacture of shaped, pyrogenic-silica-based products, e.g., an insulation body, for which, at equal density, the mechanical performance is also improved

14. Accordingly, a very important feature of the invention described and claimed in the subject application is the pressing of mixtures in a dry condition. The matrix obtained by a wet process is totally different, both texturally and structurally, from the matrix obtained by a dry

process in accordance with the invention. These lead to totally different types of products with big differences in thermal insulation performance, as demonstrated by the experiments described above.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that the statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Further declarant sayeth naught.

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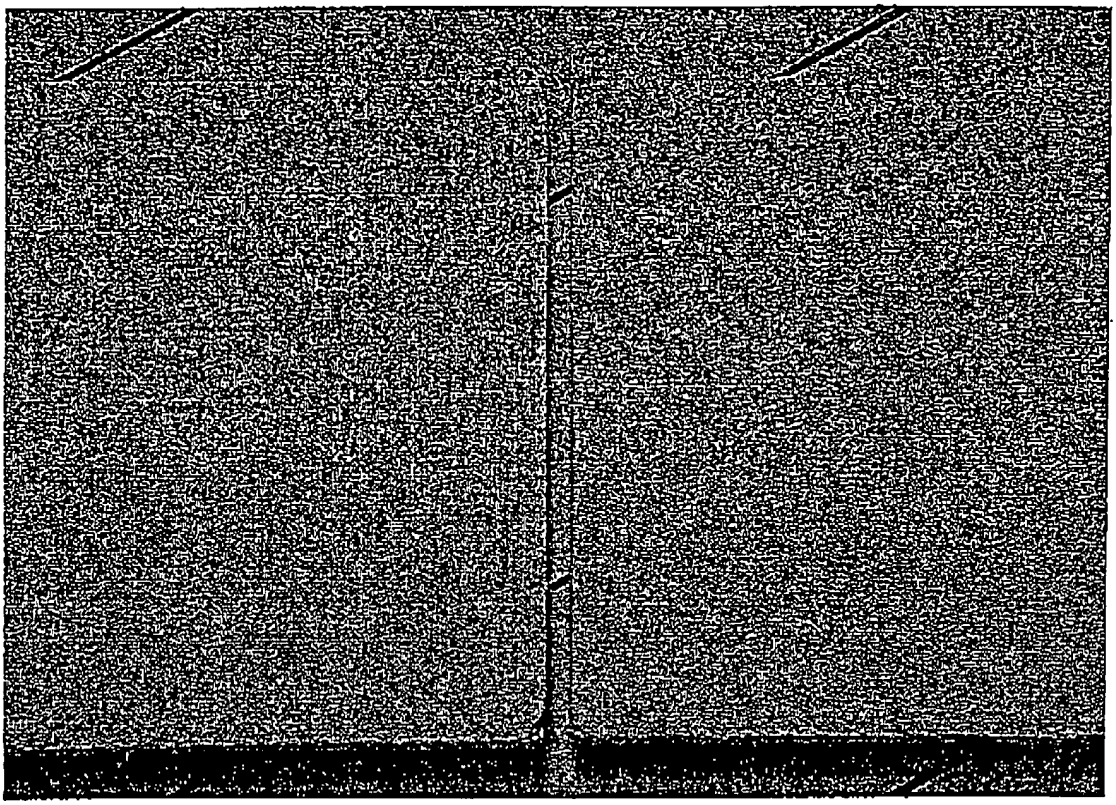
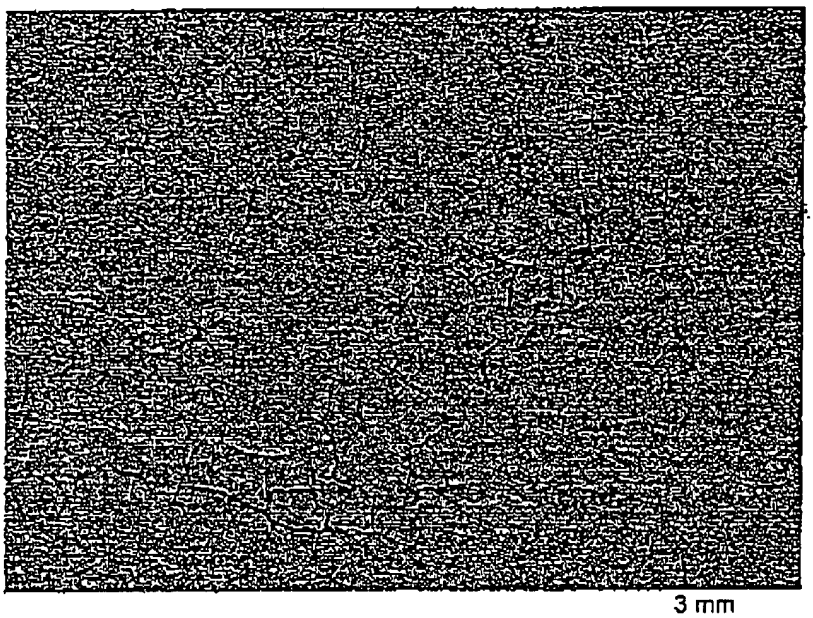
Date Octavian ANTON



Left : sample with pyrogenic silica, fibres, opacifiers and xonotlite formed by dry pressing
Right : same mixture when wet processing is applied and in which cracking is observed. The structure at the right is more clearly visible in the detailed figure below.

Fig.1 : Differences in structure between samples formed by dry pressing (left side) and wet pressing (right side).



Detail at larger magnification showing the structure of a mixture with pyrogenic silica, fibres, opacifier and xonotlite, when the latter is added as a slurry.

Fig.2

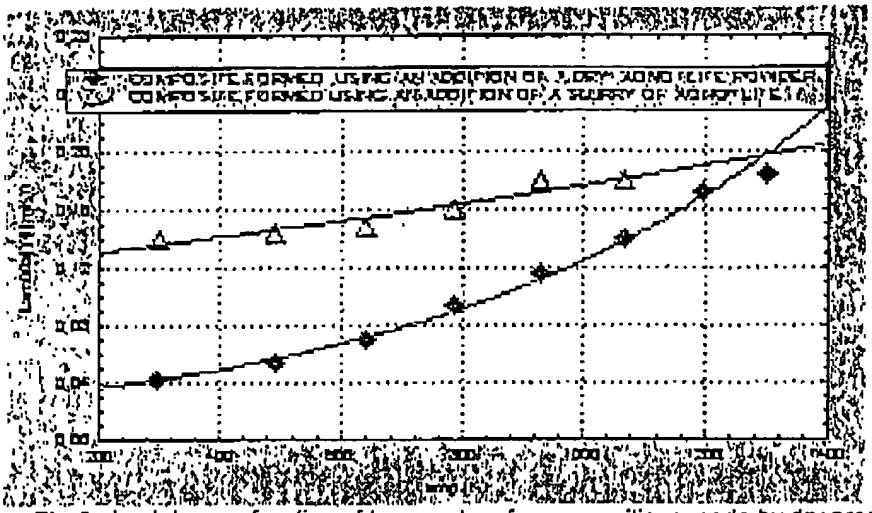
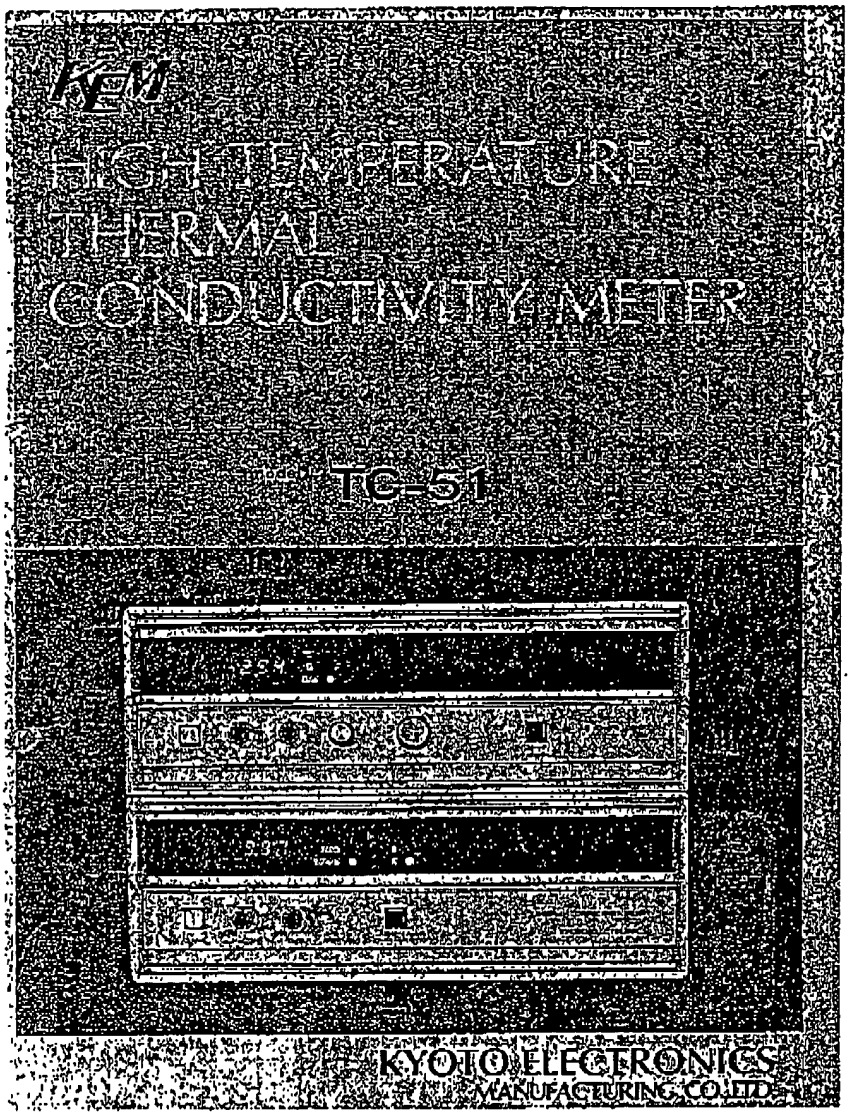
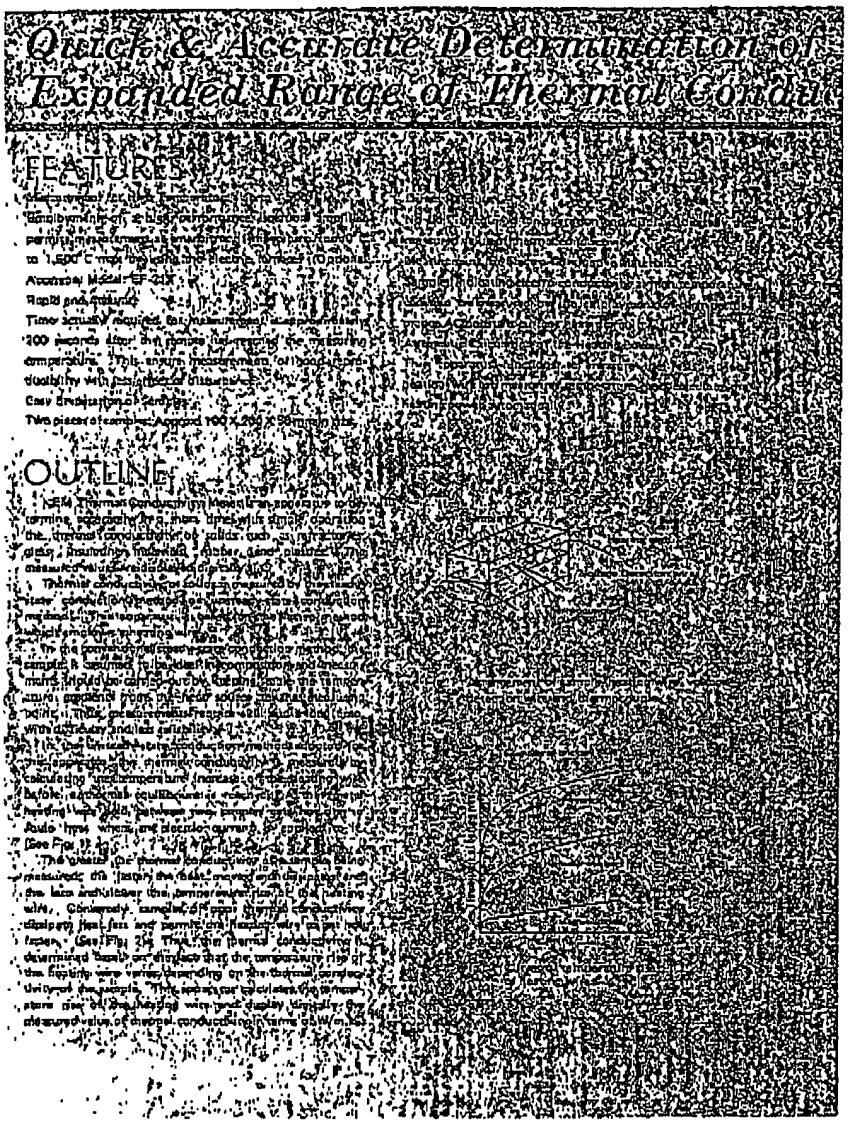


Fig.3 : lambda as a function of temperature for compositions made by dry pressing (green curve) and wet processing (magenta-curve).

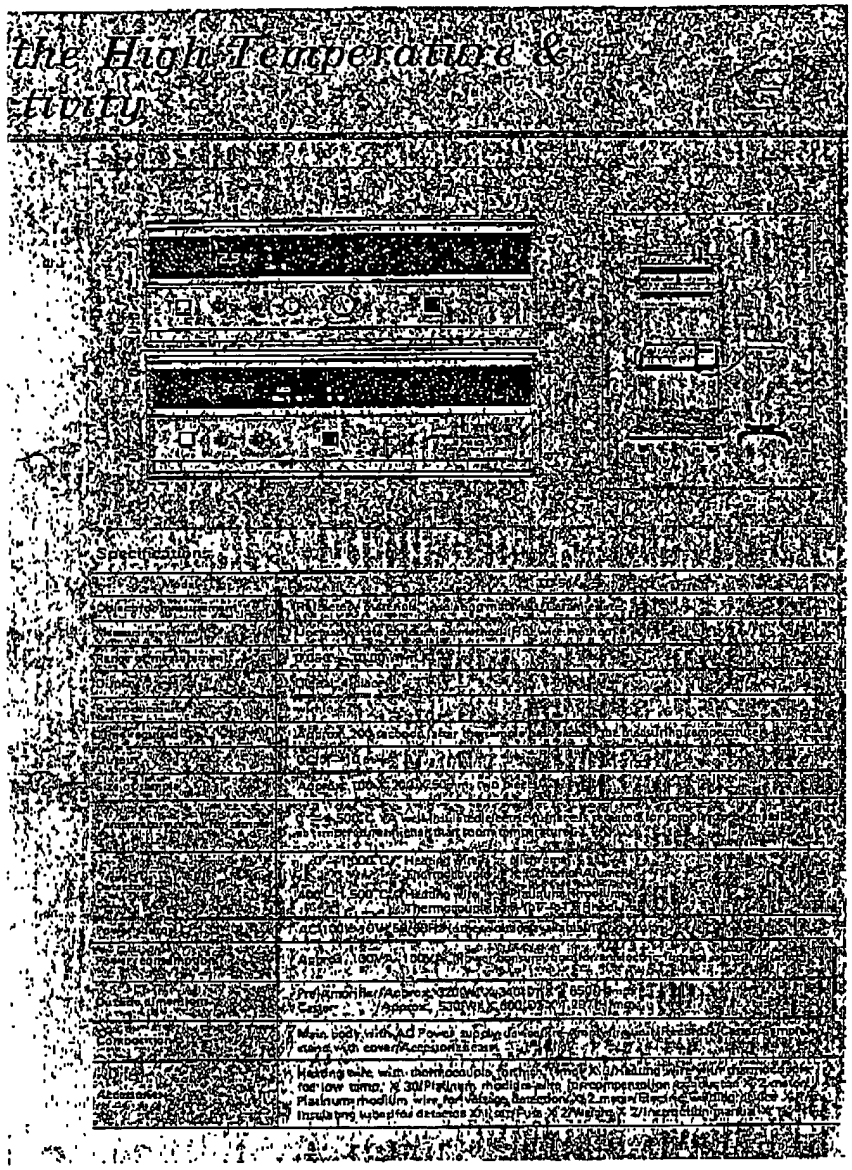




Quick & Accurate Determination of
Expanded Range of Thermal Conduc

FEATURES

OUTLINE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